


## EXPRESS MAILING CERTIFICATE NO. EJ749894952US

Form PTO-1390 (REV 10-96)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER  <b>81000.3000</b>
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371			U.S. APPLICATION NO. (If known, file 37 CFR 1.5) <b>09/403192</b>
INTERNATIONAL APPLICATION NO.  <b>PCT/EP98/01870</b>	INTERNATIONAL FILING DATE  <b>31 March 1998</b>	PRIORITY DATE CLAIMED  <b>15 April 1997</b>	
TITLE OF INVENTION  <b>PLANT PROTECTION AGENTS</b>			
APPLICANT(S) FOR DO/EO/US  <b>Jacki Müller, Achim Zöllkau, Vera Pohl, Ewa Bednarska and Rainer Süssman</b>			
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:			
<ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input type="checkbox"/> This a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li><input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19<sup>th</sup> month from the earliest claimed priority date.</li> <li><input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li><input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li><input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li><input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)), including Figures 1-3.</li> <li>Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> <li><input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li><input type="checkbox"/> have been transmitted by the International Bureau.</li> <li><input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li><input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li><input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li><input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)) (unsigned).</li> <li><input checked="" type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>			
Items 11 to 16 below concern document(s) or information included:			
<ol style="list-style-type: none"> <li><input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li><input checked="" type="checkbox"/> An assignment to Stefes Agro GMBH (unsigned).</li> <li><input checked="" type="checkbox"/> A preliminary amendment.</li> <li><input type="checkbox"/> A substitute specification.</li> <li><input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li><input type="checkbox"/> Other items of information: <ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> A copy of International Search Report.</li> </ol> </li> </ol>			

U.S. APPLICATION NO. (if known, see 37 CFR 1.5) <div style="font-size: 1.5em; font-weight: bold; text-align: center;">09/403192</div>	INTERNATIONAL APPLICATION NO. <div style="text-align: center;">PCT/EP98/01870</div>	ATTORNEY'S DOCKET NUMBER <div style="text-align: center;">81000.3000</div>	
17. <input checked="" type="checkbox"/> The following fees are submitted:  <b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1) - (5)):</b> Search Report has been prepared by the EPO or JPO (small entity). . . . . \$930.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) . . . . . \$700.00  No international preliminary examination fee paid to ISPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) . . . . . \$ 770.00  Neither international preliminary examination fee (37 CFR 1.482) nor International search fee (37 CFR 1.445(a)(2)) paid to USPTO . . . . . \$1040.00  International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) . . . . . \$ 96.00  <div style="text-align: right;"><b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b></div>		<b>CALCULATIONS PTO USE ONLY</b>	
		<b>\$ 840.00</b>	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).			
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	13-20 =	0	x \$ 22.00
Independent claims	2- 3 =	0	x \$ 82.00
MULTIPLE DEPENDENT CLAIMS (if applicable)			+ \$270.00
<b>TOTAL OF ABOVE CALCULATIONS =</b>		<b>\$ 840.00</b>	
Reduction of ½ for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).		<b>\$ -420.00</b>	
<b>SUBTOTAL =</b>		<b>\$ 420.00</b>	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).			
<b>TOTAL NATIONAL FEE =</b>		<b>\$ 420.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		<b>\$</b>	
<b>TOTAL FEES ENCLOSED =</b>		<b>\$ 420.00</b>	
		<b>Amount to be:</b>	
		<b>Refunded</b>	<b>\$</b>
		<b>Charged</b>	<b>\$</b>
a. <input checked="" type="checkbox"/> A check in the amount of <b>\$420.00</b> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. 19-3555 in the amount of \$ _____ to cover the above fees. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 19-3555. A duplicate copy of this sheet is enclosed.			
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b) must be          filed and granted to restore the application to pending status.</b>			
<b>SEND ALL CORRESPONDENCE TO:</b>			
<b>Ann W. Speckman</b> <b>LAW OFFICES OF ANN W. SPECKMAN</b> 2601 Elliott Avenue, Suite 4185 Seattle, Washington 98121 206.269.0565 direct 206.269.0563 facsimile		 <b>Jim Klaniacki</b> Registration Number 38,207 October 14, 1999	

424 Rec'd PCT/PTO 13 JAN 2000

CERTIFICATE OF MAILING

I hereby certify that this paper or fee is being deposited with the United States Postal Service as first class mail on the date indicated below and is addressed to the Assistant Commissioner for Patents, Box PCT Washington, D.C. 20231.  
Date: January 6, 2000

Jim Klaniecki

Attorney Docket No. 81000.3000  
UTILITY PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

U.S. Patent Appln. No. : 09/403,192  
Filing Date : October 14, 1999  
  
International Application No. : PCT/EP98/01870  
International Filing Date : 31 March 1998  
  
Inventor : Jacki Müller, Achim Zöllkau, Vera Pohl, Ewa Bednarska and Rainer Süßmann  
  
For : PLANT PROTECTION AGENTS

Adjustment date: 02/28/2000 WCLAYBRO 0015197200  
01/24/2000 UNWALKER 00000162 09403192  
01 FC:154 Box PCT -130.00 OP  
Washington, D.C. 20231

01/24/2000 UNWALKER 00000162 09403192  
01 FC:154 130.00 OP

Repln. Ref: 02/28/2000 WCLAYBRO 0015197200  
DAH:193555 Name/Number:09403192  
FC: 704 \$65.00 OP  
**TRANSMITTAL LETTER - RESPONSE TO NOTIFICATION**  
**OF MISSING REQUIREMENTS UNDER 35 U.S.C. 371**

TO THE ASSISTANT COMMISSIONER FOR PATENTS:

Dear Sir:

In response to the Notification of Missing Requirements under 35 U.S.C. 371 dated December 10, 1999 in connection with the subject patent application, enclosed for filing are the following:

02/28/2000 WCLAYBRO 00000112 09403192

01 FC:254 [X] Copy of Form PCT/DO/EO/905, Notification of Missing Requirements.  
[X] Executed Combined Declaration and Power of Attorney. 65.00 OP

- [X] A check in the amount of \$170.00 in total payment of the Surcharge for Late Filing of Declaration under 37 CFR 1.492(e) and the Assignment Recordation Fee..
- [ ] Please charge our Deposit Account No. 19-3555 in the amount of \$\_\_\_\_\_.
- [X] The Commissioner is hereby authorized to charge any additional fees which may be required in connection with filing of these papers, or credit overpayment, to Account No. 19-3555. A duplicate copy of this sheet is enclosed.

Respectfully submitted,

By: 

Jim Klaniecki

Registration No. 38,207

LAW OFFICES OF ANN W. SPECKMAN  
2601 Elliott Avenue, Suite 4185  
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Telephone: (206) 269-0565  
Attorney Docket No. 81000.3000

PATENT APPLICATION

Applicants : Jacki Müller, Achim Zöllkau, Vera Pohl, Ewa  
Bednarska and Rainer Süßmann  
Application No. :  
Filed : Herewith  
For : **PLANT PROTECTION AGENTS**  
Attorney Docket No. : 81000/3000

**VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY  
STATUS (37 CFR 1.9(f) and 1.27(c))--SMALL BUSINESS CONCERN**

I hereby declare that I am

☐ the owner of the small business concern identified below.

☒ an official of the small business concern empowered to act on behalf of  
the concern identified below.

NAME OF CONCERN: STEFES AGRO GMBH

ADDRESS OF CONCERN: Ottostrasse 5  
D-50170 Kerpen  
GERMANY

I hereby declare that the above-identified small business concern  
qualifies as a small business concern as defined in 13 CFR 121.3-18, and reproduced in 37  
CFR 1.9(d), for purposes of paying reduced fees under Section 41(a) and (b) of Title 35,  
United States Code, in that the number of employees of the concern, including those of its  
affiliates, does not exceed 500 persons. For purposes of this statement, (1) the number of  
employees of the business concern is the average over the previous fiscal year of the  
concern of the persons employed on a full-time, part-time or temporary basis during each  
of the pay periods of the fiscal year, and (2) concerns are affiliates of each other when  
either, directly or indirectly, one concern controls or has the power to control the other, or  
a third party or parties control or have the power to control both.

I hereby declare that rights under contract or law have been conveyed to  
and remain with the small business concern identified above with regard to the invention,  
entitled **PLANT PROTECTION AGENTS** by inventors Jacki Müller, Achim Zöllkau,  
Vera Pohl, Ewa Bednarska and Rainer Süßmann described in:

☒ the specification filed herewith.  
☐ application no. filed on .  
☐ patent no. , issued .

81000.3000

If the rights held by the above-entitled small business concern are not exclusive, each individual, concern or organization having rights to the invention is listed below\* and no rights to the invention are held by any person, other than the inventor, who could not qualify as a small business concern under 37 CFR 1.9(d) or by any concern which would not qualify as a small business concern under 37 CFR 1.9(d) or a nonprofit organization under 37 CFR 1.9(e).

\*NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27.)

FULL NAME:

ADDRESS:

☐ INDIVIDUAL    ☐ SMALL BUSINESS CONCERN    ☐ NONPROFIT ORGANIZATION

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b).)

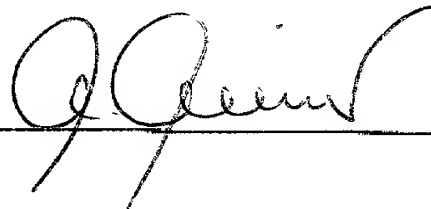
I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

NAME OF PERSON SIGNING: Manfred WEISER

TITLE OF PERSON OTHER THAN OWNER: Managing Director

ADDRESS OF PERSON SIGNING: Weimarische Strasse 25, D-10715, Berlin, Germany

SIGNATURE



DATE 6 October 1999

81000.3000

09/403122

514 733d PCT/PTO 14 OCT 1999

**CERTIFICATE OF EXPRESS MAILING**

"Express Mail" Mailing Label No. EJ749894952US

Date of Deposit: October 14, 1999

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 C.F.R. 1.10 on the date indicated above and is addressed to the DO/EO/US - FEE, U.S. Department of Commerce Patent and Trademark Office, Washington, D.C. 20231.

Jim Klaniacki

Attorney Docket No. 81000.3000  
UTILITY PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of **Jacki Müller, Achim Zöllkau, Vera Pohl, Ewa Bednarska and Rainer Süssman**

International Appln No. : PCT/EP98/01870  
International File Date : 31 March 1998  
Priority Date : 15 April 1997  
For : **PLANT PROTECTION AGENTS**  
Date : October 14, 1999

**PRELIMINARY AMENDMENT**

U.S. Department of Commerce Patent and Trademark Office  
ATTN: DO/EO/US  
Washington, D.C. 20231

Dear Sir:

Kindly amend the subject patent application as follows prior to calculation of the filing fee:

**IN THE SPECIFICATION:**

On page 1, above the first paragraph, insert the title "PLANT PROTECTION AGENTS".

On page 1, after the title of the application, insert the heading: "Technical Field of the Invention".

On page 1, after the second paragraph and before the line beginning "It is very common", insert the heading --Background of the Invention--.

On page 3, after the second paragraph and before the line beginning "It was the object of the present invention", insert the heading: --Summary of the Invention--.

On page 3, after the fourth full paragraph ending with the line "preparations and suspension concentrates," insert the heading: --Brief Description of the Drawings--.

On page 3, after the inserted heading --Brief Description of the Drawings--, insert the following description of the drawings:

"Fig. 1 shows the correlation of activity (%) and silica gel content (g/l) for a phenmedipham/desmedipham (PMP/DMP) suspension concentrate mixture.

Fig. 2 illustrates a comparison of activity of two crop protection compositions in various plant species.

Fig. 3 shows a comparison of activity of the crop protection compositions at different application rates.

On page 3, before the text of the last paragraph, before the line beginning "The silicic acid used", insert the heading: --Detailed Description of the Invention--.

### **IN THE CLAIMS:**

In Claim 3, lines 1 and 2, delete "any of" and change "Claims 1 or 2" to --Claim 1--;

In Claim 4, lines 1 and 2, delete "any of" and change "Claims 1 to 3" to --Claim 1--;

In Claim 5, lines 1 and 2, delete "any of" and change "Claims 1 to 4" to --Claim 1--;

In Claim 6, lines 1 and 2, delete "any of" and change "Claims 1 to 5" to --Claim 1--;

In Claim 7, lines 1 and 2, delete "any of" and change "Claims 1 to 6" to --Claim 1--;

In Claim 8, lines 1 and 2, delete "any of" and change "Claims 1 to 7" to --Claim 1--;

In Claim 9, lines 1 and 2, delete "any of" and change "Claims 1 to 8" to --Claim 1--;

In Claim 10, lines 1 and 2, delete "any of" and change "Claims 1 to 9" to --Claim 1--;

In Claim 11, lines 1 and 2, delete "any of" and change "Claims 1 to 10" to --Claim 1--;

and


In Claim 12, line 2, delete "any of" and change "Claim 1 to 11" to --Claim 1--;



### REMARKS

The specification has been amended to insert the headings and brief description of the drawings required according to U.S. Patent practice. The claims have been amended to reduce the total number of claims by reducing the multiple dependent claims. Favorable consideration of the claims, as amended, is requested.

Respectfully Submitted,

By:   
Jim Klamiecki  
Registration No. 38,207

LAW OFFICES OF ANN W. SPECKMAN  
2601 Elliott Avenue, Suite 4185  
Seattle, Washington 98121  
Telephone: 206.269.0565  
October 14, 1999

The present invention provides crop protection compositions comprising active compounds having foliar or systemic action which make it possible to widen the activity spectrum and the activity by employing a surfactant/silica gel mixture.

In the terminology of the present application, crop protection compositions are ready-to-use preparations of active compounds which are directed in particular against animal, microbial or plant pathogens of useful or crop plants.

It is very common and has been known for a long time to add colloidal silicic acids in the field of the formulations of crop protection agents. The use is described sufficiently in the corresponding literature.

The use known from the prior art is limited to the use of the silica gels as so-called "inert" cocomponents in crop protection preparations. Thus, for example, the use of silica gels as auxiliaries for preparing dispersible suspensions is described in Büchel "Pflanzenschutz und Schädlingsbekämpfungsmittel", 1977, page 198. In "Surfactants in Agrochemicals" 1995, page 155, Tadros describes the use of silica gels as additives for suspension concentrates to prevent sedimentation and caking.

The use of silica gel as filler is described in Foy and Pritchard in "Pesticide Formulation and Adjuvant Technology", 1996, page 85.

In Römpp, "Chemie Lexikon", 1995, page 2233, under the entry "Kieselgele", likewise only the properties and uses which are already known, inter alia as thixotropic agent, are described.

An increase in activity or a widening of the activity spectrum of, for example, herbicides having foliar action on weeds outside the known area of use on addition of silica gels has not been described and was also not to be expected.

To improve the activity of known crop protection agents, the prior art proposes specific additives or specific cocomponents developed for this purpose. In general, mineral oils and vegetable oils and their derivatives, and also specific surfactants, for example polysaccharides, ethoxylated triglycerides, ethylene oxide or propylene oxide copolymer adducts with ethylenediamine as central starter unit, polyethoxylated fatty acids and amides thereof and also ethoxylated alkylaryl alcohols having typically 6 - 10 oxyethylene units are employed here.

The amount of such additives or bioactivators that is required varies between 100 g/l of ready-to-use preparation to from 2 to 3 l per ha, for example in the case of the mineral and vegetable oils as tank mix partners of the herbicides having foliar action.

It was not possible with any of these products to extend the activity spectrum of, for example, active compounds employed as herbicides to other weeds and to improve the activity at the same time.

Recent studies have shown that the use of aqueous suspension concentrates generally offers advantages compared to emulsions. Thus, the use of solvents which are toxicologically and ecotoxicologically objectionable, such

as, for example, isophorone, cyclohexanone, xylenes and acetophenone, can be dispensed with.

Moreover, the concentration of the active compounds can be considerably increased (cf. DE 43 29 974). These advantages reduce, for example, packaging, transport and also storage costs considerably.

It was the object of the present invention to improve the activity spectrum and the activity of known active crop protection agents and to provide novel, more effective crop protection compositions.

This object was surprisingly achieved by the features of the main claim. Preferred embodiments are characterized in the subclaims.

In addition to the expected effects when rendering suspensions thixotropic and on use as a filler in powders and granules, the simultaneous use of the inorganic adsorbents selected according to the invention, preferably the use of silicic acid, aluminosilicates and/or aluminium oxides in combination with specific surfactants in powders, granules and aqueous suspension concentrates of active compounds having foliar or systemic action resulted in an unexpected increase in the activity potential and in a widening of, in particular, the weed application spectrum, compared with conventionally formulated emulsion preparations and suspension concentrates.

The silicic acid used can be colloidal silicic acid, generally referred to as silica gel, but also the so-called pyrogenic silicic acid, generally referred to as Aerosil. Aluminium oxide in all its modifications and hydrated forms and aluminosilicates, in particular

the specific surfactants selected according to the invention likewise exhibited a surprising activity-increasing effect and resulted in a widening of the application spectrum. Preference is given to using silica gel. The inorganic adsorbents selected according to the invention can be added in proportions of from 0.5 to 25.0% by weight, preferably in proportions of from 2.0 to 15.0% by weight, to the novel crop protection compositions according to the invention.

The surfactants to be employed according to the invention are selected from the group of ethoxylated C<sub>6</sub> to C<sub>20</sub> alcohols, preferably C<sub>8</sub>-C<sub>16</sub>, of the ethylene/propylene oxide block copolymers, if appropriate comprising ethylene diamine as starter unit, and of the alkyl ether sulphates and their combinations. It is also possible to employ ethoxylated castor oil. Preference is given to using ethoxylated tridecanols having from 5 to 13 ethylene oxide units, sodium, potassium or ammonium alkyl ether sulphates, in particular sodium, potassium or ammonium fatty alcohol diglycol ether sulphates or mixtures of these surfactants. Particular preference is given to using the surfactants Volpo T/785, Volpo T/10, Genapol LRO, Emulsogen, Pluronic types or mixtures of these surfactants, which are known per se. The proportion of surfactant in the crop protection compositions according to the invention is from 5.0 to 40.0% by weight, preferably from 7 to 35% by weight, particularly preferably between 10.0 and 25% by weight.

The sum of the proportions of adsorbents selected according to the invention and surfactants selected according to the invention is from 5.5 to 45.0% by weight, preferably from 15.0 to 25.0% by weight.

As herbicidally active compounds, the novel crop protection compositions according to the invention comprise herbicides which have both foliar action and soil and foliar action from the groups of the urea derivatives, the carbamates, biscarbamates, diphenyl ethers, pyridolylacetic acid derivatives, pyridazines, triazines, triazinones, uracils, sulphonylureas, benzofuran derivatives and also glufosinate and glyphosate, and their salts or esters, and also mixtures of some selected active compounds. Preference is given to using phenmedipham (PMP), desmedipham (DMP), metamitron and/or ethofumesate.

Suitable fungicidally active compounds are morpholines, azols, phthalimides or piperidines, suitable insecticides are pyrethroids, carbamates or organophosphates, and the possible salts or esters and also mixtures of selected abovementioned active compounds. The proportion of active compound in the crop protection compositions according to the invention is from 5.0 to 75.0% by weight, preferably from 15.0 to 55.0% by weight.

In addition to these components, the crop protection composition according to the invention may comprise auxiliaries and/or carriers known per se which are customarily used for suspension concentrates. Antifreeze agents, stabilizers, antifoams, wetting agents and dispersants and also, if appropriate, other fillers may be mentioned by way of example. Reference may also be made here to DE 43 29 974. The wetting agent and dispersant can be selected, for example, from the groups of the phosphated di- or tristyrenephenol ethoxylates in the phosphate form and/or of the lignin sulphonates. Preference is given to using ethoxylated tristyrenephenol phosphates, sodium, potassium, calcium, magnesium, zinc or ammonium lignin sulphonates, in particular mixed calcium/magnesium/zinc lignin sulphonate salts, or mixtures of these agents.

Particular preference is given to Tensiofix LX Special, Soprophor FL, Soprophor FL 60 or mixtures of these agents.

The crop protection compositions according to the invention are prepared by processes known per se, for example the wet milling principle. To this end, the individual components (active compounds and auxiliaries) are finely ground with an appropriate amount of water in a suitable apparatus, for example a bead mill.

The components are preferably adjusted to a degree of fineness of from 0.5 to 20  $\mu\text{m}$ , in particular to a degree of fineness of < 10  $\mu\text{m}$ . In the case of solid preparations (granules or powders), use is likewise made of methods corresponding to the prior art.

In the formulation form/appearance of a suspension concentrate, the crop protection compositions according to the invention exhibit, in addition to the known toxicological and ecotoxicological advantages, surprisingly an additionally increased biological potential in the control of weeds which as yet has not been able to be controlled using equivalent application rates of known preparations.

Thus, compositions prepared using the adsorbent/surfactant combination according to the invention make it possible for the first time to employ crop protection agents from the group of the carbamates and biscalbamates for controlling camomile species, for example *Matricaria chamomilla* (MATCH) and *Matricaria inodora* (MATIN). Hitherto, it has not been possible to control camomile species satisfactorily, either using the active compound desmedipham, or using the active compound phenmedipham, or using a 1:1 mixture or a combination of the abovementioned

active compounds with ethofumesate (cf. the examples below). Satisfactory control has hitherto only been possible by using further herbicides (for example metamitron).

Furthermore, it was possible to reduce the amount of composition applied by from 10 to 20% at the same activity, for example, by employing the crop protection compositions according to the invention from the field of the triazinones, for example in the cultivation of sugar beet, as shown by the example below.

The table shows the scores obtained in a herbicide trial in sugar beet. A conventional metamitron SC was compared, at an equivalent application rate in l/ha, but with reduced amounts of active compound per ha, with a metamitron SC according to the invention.

(6.5 l/ha x 700 g/l = 4550 g/ha compared with 6.5 l/ha x 500 g/l = 3575 g/ha corresponding to -21.4%)

Preparation	l/ha	CHEAL	VIOAR	POLSS	MATCH	GALAP	MEAN
metamitron 700 g/l SC	6.5	98	92	90	100	73	91
metamitron SC according to the invention, 550 g/l	6.5	100	88	96	100	80	93

Table 1

The combination of two active compounds having foliar action, i.e. phenmedipham and desmedipham, whose known activity spectra are limited to broad-leaved weeds, with insufficient activity against camomile, showed, at an identical application rate of active compound per ha, a considerably improved activity compared to an emulsion



The following examples and recipes are intended to illustrate the invention, without limiting it.

## Suspension concentrate A:

	g/l
desmedipham	160
phenmedipham	160
Soprophor FL 60	25
Volpo T/7 85	100
Genapol LRO	30
Tensiofix LX Spezial	5
silica gel SM 614	40
antifreeze agent	40
stabilizer	1
antifoam	5
water	ad 1 1

## Suspension concentrate B:

	g/l
desmedipham	35
phenmedipham	100
ethofumesate	200
Pluronic	<u>55</u>
Genapol LRO	150
silica gel SM 614	35
antifreeze agent	60
stabilizer	0.5
antifoam	5
water	ad 1 1

X  
X

## Suspension concentrate C:

	g/l
desmedipham	320
Soprophor FL	30
Volpo T/7 85	135
Genapol LRO paste	15
Tensiofix LX Spezial	3
silica gel SM 514	37
antifreeze agent	45
stabilizer	0.5
antifoam	5
water	ad 1 1

## Suspension concentrate D:

	g/l
phenmedipham	320
Soprophor FL 60	45
Volpo T/10	120
Genapol LRO	45
Tensiofix LX Spezial	5
silica gel SM 614	45
antifreeze agent	40
stabilizer	1.0
antifoam	7
water	ad 1 1

antifreeze agent	45	antifreeze agent	40
stabilizer	0.5	stabilizer	1.0
antifoam	5	antifoam	7
water	ad 1 l	water	ad 1 l

Suspension concentrate E:

	g/l
metamitron	550
Hoechst LFS	35
Genapol LRO	40
Volpo T/10	120
silica gel Tixosil 38A	37
antifreeze agent	35
antifoam	5
stabilizers	1
water	ad 1 l.

Example 1:

Correlation of activity (%) and silica gel content (g/l)  
for a PMP/DMP mixture:

The effect of the addition of silica gel on the activity potential of a crop protection composition becomes evident when comparing a basic recipe without and with varying silica gel content. A phenmedipham/desmedipham suspension concentrate (PMP/DMP) having varying silica gel content was compared with a phenmedipham/desmedipham emulsion concentrate Betanal<sup>®</sup> AM 11 EC from Hoechst Schering AgrEvoGmbH (Betanal AM 11 comprises PMP/DMP in a ratio of 1:1) without silica gel.

The results are shown in Table 2 and Figure 1.

Correlation of activity (%) and silica gel content (g/l) for a PMP/DMP mixture at the same application rate of active compounds/ha				
Silica gel content in a suspension according to the invention (series 1)	0	20	30	40
Activity %	56.6	85.8	90.8	94.2
*Betanal AM 11 EC without silica gel (series 2)	0	0	0	0
Activity %	75	75	75	75

Table 2

Example 2: Comparison of the activity of two crop protection compositions

A crop protection composition according to the invention (suspension concentrate B; SC B) which comprised 320 g/l of desmedipham was compared with a crop protection composition known from the prior art (Betanal® AM from Hoechst Schering AgrEvo GmbH; Betanal AM comprises only desmedipham; EC), which comprised 160 g of desmedipham/l. 0.75 l of the suspension concentrate according to the invention (SC B)/ha and 1.5 l of Betanal AM (EC)/ha were applied (this corresponds to equivalent amounts/ha). The results are shown in Tab. 3 and Fig. 2.

Comparison of the activity of a suspension according to the invention with *Betanal AM							
Type	Activity [%]/species						
	GALAP	MATCH	MATIN	STEME	CHEAL	POLPE	Mean
SC B	30	60	35	55	95	20	49.2
EC	40	30	20	0	100	20	35

GALAP: Galium aparine

MATCH: Matricaria chamomilla

MATIN: Matricaria inodora

STEME: Stellaria media

CHEAL: Chenopodium album

POLPE: Polygorium persicara

Table 3

Example 3: Comparison of the activity at different application rates:

The substantially improved activity of the crop protection compositions according to the invention becomes evident when the relative fresh weights of the weeds are compared. The suspension A according to the invention (SC A) was compared with Betanal<sup>®</sup> AM 11 from AgrEvo which comprised DMP and PMP in a ratio of 1:1.

Fresh weight is defined as the mass of the above-ground parts of the plants.

Relative fresh weight is defined as the mass of the above-ground parts of the plants of a treated sample in comparison with the mass of an untreated control sample (=100%).

The results are shown in Table 4 and Figure 3.

<u>Comparison of the relative fresh weights at two different application rates</u>								
Product	Appli- cation rate [l/ha]	BEAVA	POLPE	MATCH	MATIN	STEME	CHEAL	GALAP
Betanal AM 11	4	67	4	23	15	27	0	1
SC A	2	55	8	0	0	1	0	0
Betanal AM 11	2	90	31	54	38	50	0	6
SC A	1	73	14	1	2	13	0	6

GALAP: Galium aparine

MATCH: Matricaria chamomilla

MATIN: Matricaria inodora

STEME: Stellaria media

CHEAL: Chenopodium album

POLPE: Polygorium persicara

BEAVA: Beta vulgaris

Table 4

#### Example 4:

Example 4 shows a comparison of the activity of approved emulsion concentrates (Betanal AM and Betanal AM 11) with suspension concentrate according to the invention having a proportion of silica gel of 40 g/l or 20 g/l at different application rates. Both the integral (6 species) and the species-specific activity is substantially increased when the composition according to the invention is employed.

Variant	Active compounds (g/l)	Appli- cation rate (l/ha)	Activity %		
			Ø* 6 species	Matin**	Match***
Standard EC Betanal ®AM11	80 + 80 DMP + PMP	1	22.6	0	0
		2	46.5	45	10
		4	75	75	50
Suspension according to the invention SC A (Code No. 8102- 005I97 comprising 40 g of silica gel/l)	160 + 160 DMP + PMP	0.5	46.6	65	55
		1	82.3	97	97
		2	94.2	100	100

Variant	Active compounds (g/l)	Appli- cation rate (l/ha)	Activity %		
			ø*		
			6 species	Matin**	Match***
Standard EC Betanal ® AM	160 DMP	1	20	0	30
		1.5	35	20	30
		2	46.7	20	40
Suspension according to the invention SC B (Code No. 8101- 006I97 comprising 20 g of silica gel/l)	320 DMP	0.5	35.8	20	60
		0.75	49	35	60
		1	62	75	70

Table 5

- \* Galium aparine, matricaria chamomilla, matricaria inodore, stellaria media, Chenopodium album and polygonum persicara
- \*\* Matricaria inodora
- \*\*\* Matricaria chamomilla
- Betanal ® registered trademark of Hoechst Schering AgrEvo GmbH

Example 5:

Comparison of approved emulsion concentrates with suspension concentrates according to the invention comprising the active compounds ethofumesate, phenmedipham and desmedipham with a proportion of silica gel of 40 g/l.

	l/ha	GALAP	MATCH	MATIN	STEME	CHEAL	POLPE
Betanal Progress comprising	1	75	90	99	75	99	80
PMP 75; DMP 25; Etho 150 g/l							
SC according to the invention comprising	0.75	75	95	100	88	100	90
PMP 100; DMP 33; Etho 200 g/l							

Table 6

Table 7 below states the type and group association of the cocomponents used.



Cocomponent	Type	Chem. group
Volpo T 7 85 and T 10	Wetting agent	Polyethoxylated alcohols
Genapol LRO	Wetting agent	Fatty alcohol diglycol ether sulphate Na salt
Tensiofix LX Special	Dispersant	Lignin sulphonate
Soprophor Fl and Fl 60	Dispersant	Ethoxylated tristyrene- phenol phosphate
Emulsogen	Wetting agent	Ethoxylated castor oil
Pluronic	Dispersant	Ethylene oxide/propylene oxide block polymer
Hoechst LFS	Dispersant	Poly(arylalkyl)phenol polyethylene glycol phosphoric ester, triethanolammonium salt

Table 7

## PATENT CLAIMS

1. Crop protection composition, formulated as powder, granules or water-based, of active compounds having foliar or systemic action, characterized in that it comprises, in the case of herbicide preparations, at least one herbicide from the groups of the urea derivatives or sulphonylureas, the carbamates, biscarbamates, diphenyl ethers, pyridolylacetic acid derivatives, pyridazines, triazines, triazinones, uracils, benzofuran derivatives, glyphosate or glufosinate, in the case of fungicide preparations, at least one fungicide from the group of the morpholines, azoles, phthalimides or piperidines, in the case of insecticide preparations, at least one insecticide from the group of the pyrethroids, carbamates or organophosphates, or possible salts or esters of the abovementioned groups of active compounds, at least one inorganic adsorbent and at least one surfactant, preferably from the group of the ethoxylated C<sub>6</sub>- to C<sub>20</sub>-alcohols, preferably C<sub>8</sub>- to C<sub>16</sub>-alcohols, the ethoxylated castor oils or the alkyl ether sulphates,

the combination of metamitron technical (I), ethofumesate technical (II), phenmedipham technical (III), alcylyl sulfonate (IV), polyoxyethylene-(6)-tridecyl ether (V), highly disperse precipitated silicic acid (VI) and kaoline W (VII) in the composition:

I	II	III	IV	V	VI	VII
[%-wt.]	[%-wt.]	[%-wt.]	[%-wt.]	[%-wt.]	[%-wt.]	[%-wt.]
50,0	6,60	6,80	20,0	5,0	7,5	ca. 4,10
45,0	5,94	6,12	20,0	7,5	7,5	ca. 7,94
45,0	5,94	6,12	20,0	10,0	10,0	ca. 2,94
50,0	6,60	6,80	20,0	7,0	7,0	ca. 2,60
50,0	6,60	6,80	20,0	7,0	ca. 9,6	0,0
23,3	6,30	6,50	15,0	5,0	5,0	38,9

and the combination of 33,93% by weight of propachlor (94%), 11,31% by weight of atrazine (95%), 4,00% by weight of Hi-Sil 233®, 1,00% by weight of kaoline, 2,80% by weight of Pluronic 105®, 1,00% by weight of Igepon T77®, 1,00% by weight of CaCl<sub>2</sub>·2 H<sub>2</sub>O, 8,00% by weight of ethylene glycol, 0,10% by weight of Corak 100® and 36,86% by weight of water being excluded.

4. Crop protection composition according to any of Claims 1 to 3, characterized in that the surfactant is selected from the ethoxylated tridecanols having from 5 to 13 ethoxy units, from the sodium, potassium or ammonium fatty alcohol dialkylglycol ether sulphates, from ethoxylated castor oil or from mixtures of these surfactants. X

5. Crop protection composition according to any of Claims 1 to 4, characterized in that the surfactant is selected from Volpo T/785, Volpo T/10, Emulsogen or Genapol LRO, or from mixtures of these surfactants. X

6. Crop protection composition according to any of Claims 1 to 5, characterized in that, in the case of the herbicidal preparations, the active compound is selected from phenmedipham, desmedipham, met amitron or ethofumesate, or of mixtures of these active compounds.

7. Crop protection composition according to any of Claims 1 to 6, characterized in that it additionally comprises customary auxiliaries and/or carriers, for example antifreeze agents, ~~stabilizers~~ antifoams and/or wetting agents and dispersants and also carriers, in customary proportions. X

8. Crop protection composition according to any of Claims 1 to 7, characterized in that the wetting agent and dispersant are selected from the phosphated di- or tristyrenes and lignin sulphonates, preferably from the ethoxylated trisstyrenephenol phosphates, or the sodium, potassium, calcium, magnesium, zinc or ammonium lignin sulphonates, in particular from the mixed calcium/magnesium/zinc lignin sulphonate salts, or mixtures of these agents.

# ABSTRACT

Crop protection compositions, formulated as powder, granules or water-based, of active compounds having foliar or systemic action are characterized in that they comprise, in the case of herbicide preparations, at least one herbicide from the groups of the urea derivatives or sulphonylureas, the carbamates, biscarbamates, diphenyl ethers, pyridolylacetic acid derivatives, pyridazines, triazines, triazinones, uracils, benzofuran derivatives, glyphosate or glufosinate, in the case of fungicide preparations, at least one fungicide from the group of the morpholines, azoles, phthalimides or piperidines, in the case of insecticide preparations, at least one insecticide from the group of the pyrethroids, carbamates or organophosphates, or possible salts or esters of the abovementioned groups of active compounds, at least one inorganic adsorbent and at least one surfactant, preferably from the group of the ethoxylated C<sub>6</sub>- to C<sub>20</sub>-alcohols, preferably C<sub>8</sub>- to C<sub>16</sub>-alcohols, the ethoxylated castor oils or the alkyl ether sulphates.

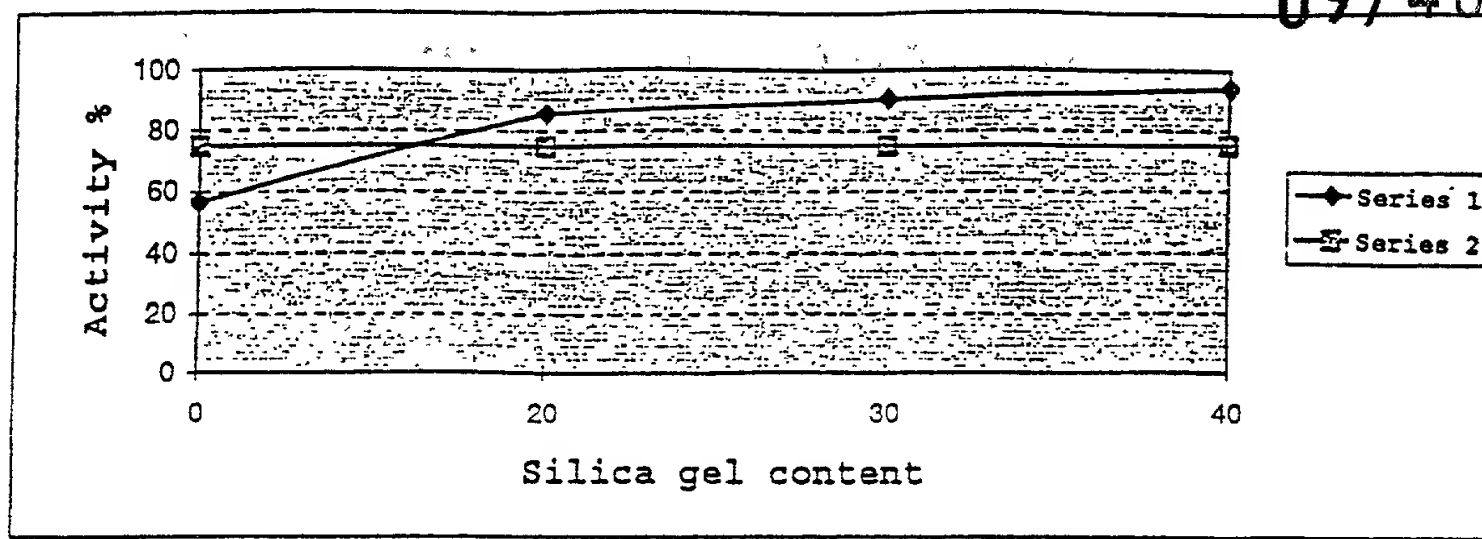


Fig. 1

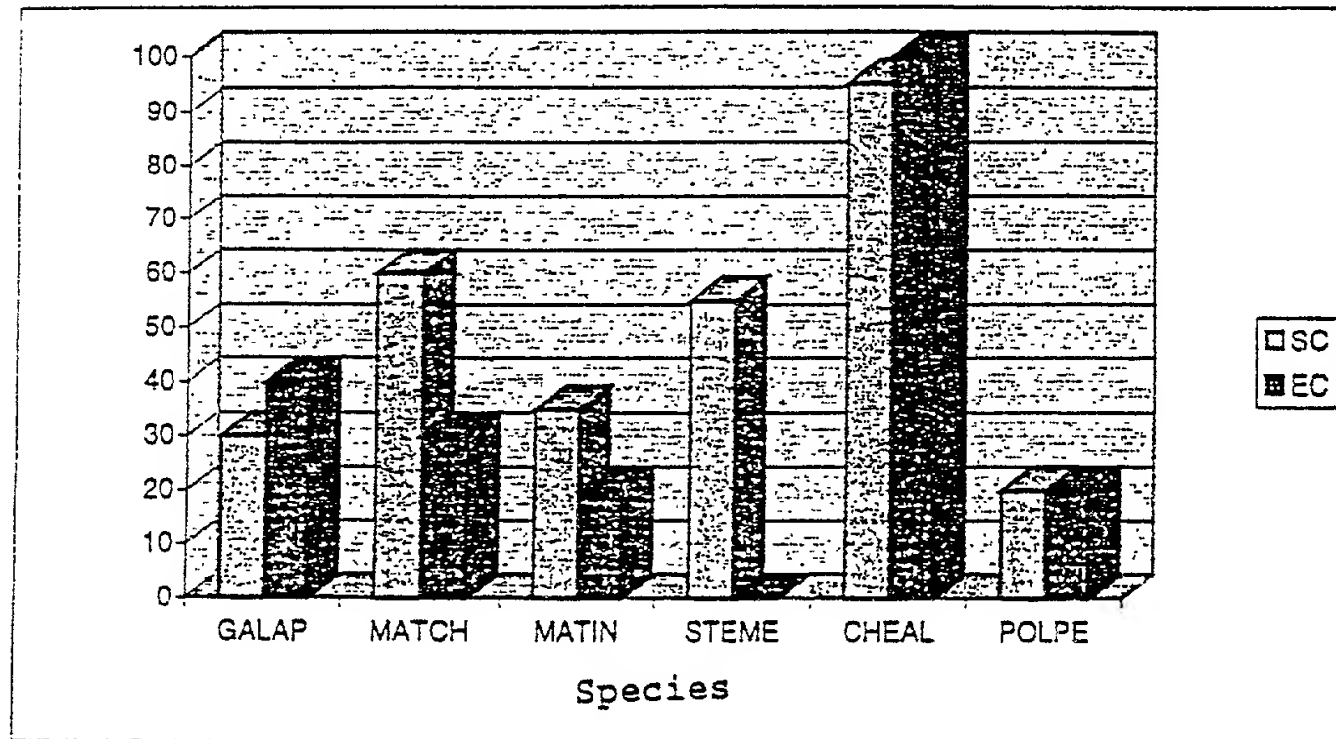


Fig. 2

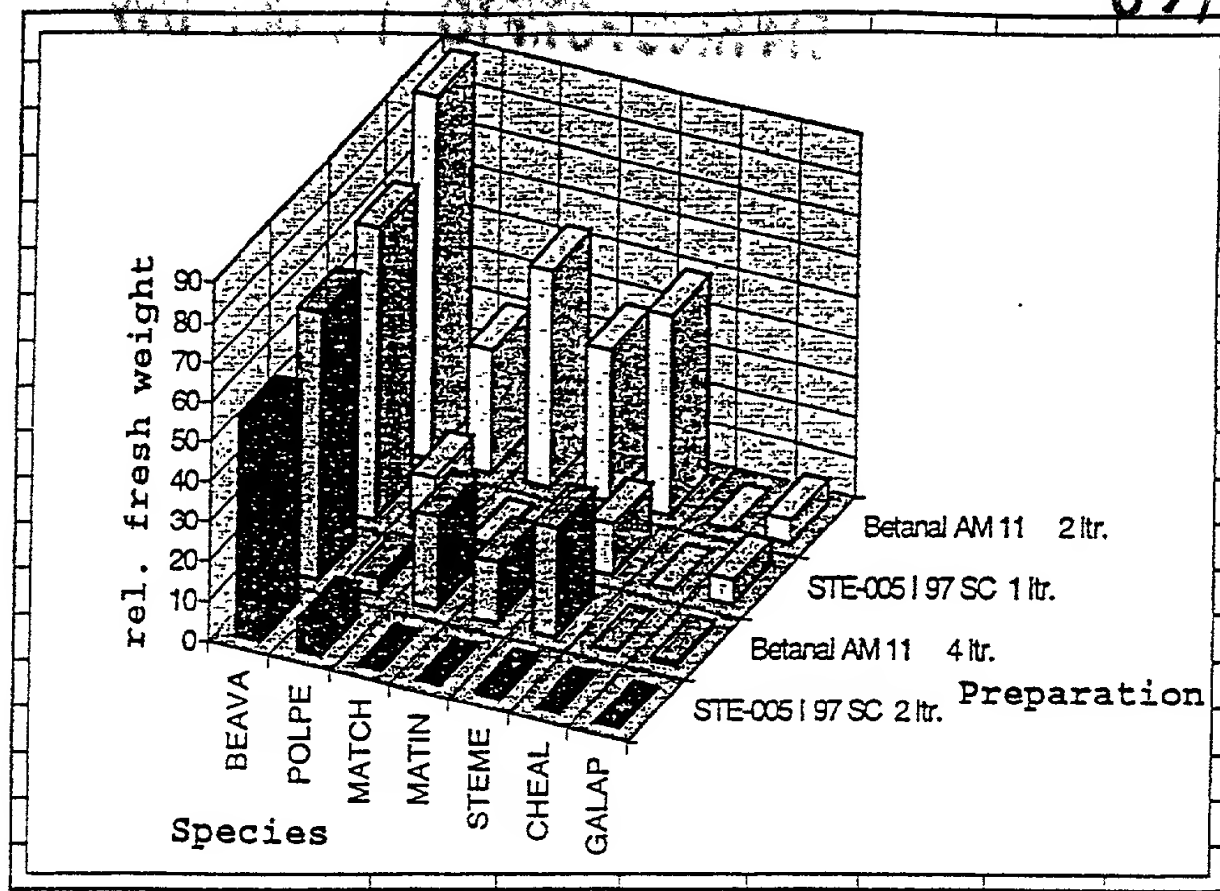


Fig. 3

**COMBINED DECLARATION AND POWER OF ATTORNEY**  
**FOR PATENT APPLICATION – MULTIPLE INVENTORS**

As below-named inventors, we, Jacki Müller, Achim Zöllkau, Vera Pohl, Ewa Bednarska and Rainer Süßmann hereby declare that:

Our residence, post office address and citizenship are as stated below next to our name.

We believe we are original and joint inventors of the subject matter which is claimed and for which a patent is sought on the invention entitled **PLANT PROTECTION AGENTS**, the specification of which

☐ is attached hereto.

☒ was filed on October 14, 1999, as Application no. 09/403,192

We hereby state that we have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

We hereby claim foreign priority benefits under Title 35, United States Code, § 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed: **PCT/EP98/01870, filed 31 March 1998; and DE 197 15 639.8, filed 15 April 1997.**

We hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, § 112, we acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of prior application and the national or PCT international filing date of this application: **PCT/EP98/01870, filed 31 March 1998.**

We hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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We hereby declare that all statements made herein of our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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